LCA Methodology

Evaluation of Ecotoxicity Effect Indicators for Use in LCIA

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Abstract

Goal, Scope and Background. The paper describes different ecotoxicity effect indicator methods/approaches. The approaches cover three main groups, viz. PNEC approaches, PAF approaches and damage approaches. Ecotoxicity effect indicators used in life cycle impact assessment (LCIA) are typically modelled to the level of impact, indicating the potential impact on 'ecosystem health'. The few existing indicators, which are modelled all the way to damage, are poorly developed, and even though relevant alternatives from risk assessment exist (e.g. recovery time and mean extinction time), these are unfortunately at a very early stage of development, and only few attempts have been made to include them in LCIA.

Methods. The approaches are described and evaluated against a set of assessment criteria comprising compatibility with the methodological requirements of LCIA, environmental relevance, reproducibility, data demand, data availability, quantification of uncertainty, transparency and spatial differentiation.

Results and Discussion. The results of the evaluation of the two impact approaches (i.e. PNEC and PAF) show both pros and cons for each of them. The assessment factor-based PNEC approach has a low data demand and uses only the lowest data (e.g. lowest NOEC value). Because it is developed in tiered risk assessment, and hence makes use of conservative assessment factors, it is not optimal, in its present form, to use in the comparative framework of LCIA, where best estimates are sought. The PAF approaches have a higher data demand but use all data and can be based on effect data (PNEC is no-effect-based), thus making these approaches non-conservative and more suitable for LCIA. However, indiscriminate use of ecotoxicity data tends to make the PAF-approaches no more environmentally relevant than the assessment factor-based PNEC approaches. The PAF approaches, however, can at least in theory be linked to damage modelling. All the approaches for damage modelling which are included here have a high environmental relevance but very low data availability, apart from the 'media recovery-approach', which depends directly on the fate model. They are all at a very early stage of development.

Conclusion, Recommendations and Outlook. An analysis of the different PAF approaches shows that the crucial point is according to which principles and based on which data the hazardous concentration to 50% of the included species (i.e. HC50) is estimated. The ability to calculate many characterisation factors for ecotoxicity is important for this impact category to be included in LCIA in a proper way. However, the access to effect data for the relevant chemicals is typically limited. So, besides the coupling to damage modelling, the main challenge within the further development and improvement of ecotoxicity effect indicators is to find an optimal method to estimate HC50 based on little data.

Keywords: Damage approaches; ecotoxicity effect indicators; hazardous concentration (HC50); life cycle impact assessment (LCIA); PAF approaches; PNEC approaches

Glossary

AF	Assessment Factor
CA	Concentration Addition
CF	Characterisation Factor
CDF	Cumulative Distribution Function
EC_{50}	Effect Concentration (50% of test organism affected)
EDIP	Environmental Design of Industrial Products
EEI	Ecotoxicity Effect Indicator
GM	Geometric Mean
HC5	Hazardous Concentration for 5% of included species
HC50	Hazardous Concentration for 50% of included species
HU	Hazard Units
LC_{50}	Lethal concentration (50% of test organism dead)
LCA	Life Cycle Assessment
LCIA	Life Cycle Impact Assessment
MET	Mean Extinction Time
msPAF	Multi-substance PAF
NOEC	No Observed Effect Concentration
OMNIITOX	Operational Models aNd Information tools for Industrial applications of eco/TOXicological impact assessments
PAF	Potentially Affected Fraction of species
PNEC	Predicted No Effect Concentration
PDF	Probability Distribution Function
PVF	Potentially Vanished Fraction of species
RA	Response Addition
SSD	Species Sensitivity Distribution
TGD	Technical Guidance Document
TMoA	Toxic Mode of Action
α	Location parameter
β	Scale parameter
γ	Slope factor

Introduction

The aim of this paper is to present a qualitative review of existing and proposed methods for ecotoxicity effect assessment including possible damage models within life cycle impact assessment (LCIA). The work described has been done as part of the OMNIITOX[1] project dealing with impact assessment of toxic chemicals within LCA and risk

[1] **OMNIITOX** is an EU-project under the Competitive and Sustainable Growth Programme, running from 2001 to 2005. OMNIITOX will facilitate decision-making regarding potentially hazardous compounds by improving methods and developing information tools necessary for impact assessment of toxic chemicals within LCA and risk assessment. Project partners are Technical University of Denmark; Leiden University, The Netherlands; University of Stuttgart, Germany; École Polytechnique Fédérale de Lausanne (EPFL), Switzerland; Chalmers University of Technology, Sweden; European Chemicals Bureau, JRC, Ispra, Italy; Volvo Technology Corporation, Sweden; Procter & Gamble EUROCOR, Belgium; Stora Enso AB, Sweden; Antonio Puig, S.A. Spain; Randa Group S.A, Spain. More information about OMNIITOX can be found at http://omniitox.imi.chalmers.se/OfficialMirror.

assessment (Molander et al. 2004). Within this project, different characterisation and selection models for LCIA of toxic chemicals were compared (Pant et al. 2004, Larsen et al. 2004), and the need for ecotoxicity effect indicators, which better meet the requirements of LCIA, was identified. The present paper documents the findings of this work, describes and evaluates the identified approaches against assessment criteria to arrive at recommendations for improvements and further developments for each of them.

An ecotoxicity effect indicator (EEI) is defined here as the 'effect part' of a characterisation factor for ecotoxicity. For example, a characterisation factor (CF) for 1 kg emitted of a given chemical can be expressed as the effect indicator (e.g. EEI = 1/PNEC) multiplied by the 'fate part' (dC), as shown in Eq. 1. A synonym for the effect indicator is effect factor as used by Pennington et al. (2004).

The methods used for EEIs within both risk assessment and LCIA fall into two main groups:

- Assessment Factor (AF) based approaches (Predicted No Effect Concentration, PNEC approaches)
- Species Sensitivity Distribution (SSD) based approaches (Potentially Affected Fraction of species, PAF approaches)

In regulatory generic risk assessment the AF approaches or PNEC approaches are used in a tiered approach to estimate a PNEC value which is combined with an estimated Predicted Environmental Concentration (PEC) resulting in a Risk Quotient (RQ=PEC/PNEC). The AFs are used conservatively and typically vary between 10 and 1000 depending on the data availability and quality of the ecotoxicity effect data (e.g. EC50 on Daphnia, fish and algae). The PNEC is estimated by dividing the lowest effect value by the chosen AF. An RQ of one or higher leads to a higher tier with a more detailed risk assessment entailing a higher data demand (EC 2003a). The SSD or PAF approaches are typically only used in higher tier risk assessment, and mostly for estimating the HC5 value (**Fig. 1**). The two approaches may be combined as the HC5 value may be used as a basis for estimating a PNEC value as described in the TGD (EC 2003a).

In LCIA the PNEC approaches are modelled to the level of potential impact on the ecosystem, i.e. a midpoint in the environmental mechanism or impact chain (ISO 2000, Udo de Haes and Lindeijer 2002). The procedure used is in principle identical to the estimation of PNEC in generic risk assessment, as described above. PAF approaches are also modelled to the level of impact but attempts to combine with damage modelling (i.e. recovery time), simply by use of general conversion factors, are known. The Eco-indicator 99 method, making use of the 'marginal PAF increase' approach (Goedkoop and Spriensma 2001a, 2001b), thus models to the damage indicator Potentially Disappeared Fraction of species (PDF of species). The PDF is in the continuation designated Potentially Vanished Fraction of species (PVF) to avoid confusion with the Probability Distribution Function (PDF) used later on in this paper. The AMI method (based on the 'average PAF increase' approach) links to damage on the same basis as the Eco-indicator 99 method but uses a different general conversion factor from PAF to PVF (Jolliet et al. 2003).

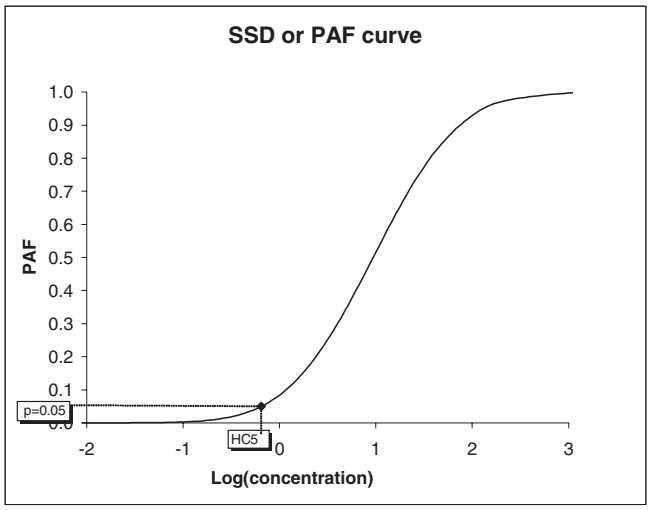

Fig. 1: Example of a species sensitivity distribution (SSD) curve or PAF curve illustrating the relationship between the environmental concentration of a toxicant and PAF, i.e. the cumulative probability of exceeding NOECs, for instance, for a certain fraction of species in the community/ecosystem in question. The concentration at which the fraction p of species are having their NOEC exceeded is denoted the hazardous concentration at level p, i.e. HCp. The concentration exceeding the NOEC for 5% (PAF=0.05) of the species, is denoted $HC5_{NOEC}$ (in this example $HC5_{NOEC}$ is $10^{-0.19}$)

As described above, a damage model is required to be able to transform a midpoint or impact indicator into an endpoint or damage indicator (e.g. expressing the results in terms of changes in biodiversity). For damage models, the following three potential approaches have been identified; they are all at an early stage of development:

- Recovery time approach (e.g. used as 'media recovery')
- Species extinction approach (Mean Extinction Time, MET)
- Changes in genetic diversity approach.

Whether attempts are made to model all the way to damage, or only to impact, the aim is that the indicator shall reflect at least some kind of relationship with potential damage to ecosystems or 'ecosystem health'. Although, as stated by Calow (1998), "we are not yet able to define general criteria of 'ecosystem health'", the biodiversity (i.e. species diversity defined as species richness) is here considered as a measure of 'ecosystem health'. Taking the state of knowledge into account, this choice seems to be the most sensible, and it is indirectly supported by Pratt and Cairns (1996), stating that "the relationship between diversity and ecological 'stability' remains elusive" but "predictions and empirical evidence suggest that species richness is a useful measure of biological diversity that responds predictably to a number of stressors".

1 PNEC-Based Impact Approaches

The PNEC approach making use of assessment factors is used in many of the existing LCIA methods, e.g. USES-LCA (Huijbregts et al. 2000) and EDIP (Hauschild et al. 1998). It is also the recommended approach in generic risk assessment according to the Technical Guidance Document (TGD) on risk assessment from the European Commission (EC 2003a).

Sub-approaches that can be identified under the overall PNEC approach include:

- PNEC based solely on acute ecotoxicity data (EC_{50}s)
- PNEC based on chronic ecotoxicity data, if possible (No Observed Effect Concentrations, NOECs)
- PNEC based on $HC5_{NOEC}$ from SSDs

Even though PNEC is based on acute data in many estimations (because of data lack), chronic data is typically preferred when available. The PNEC approach based on an $HC5_{NOEC}$ value overlaps with the PAF approaches because the PNEC value in this case is estimated as the hazardous concentration to 5% of included species ($HC5_{NOEC}$), i.e. the concentration where the NOEC value is exceeded for 5% of the species in the SSD (van Straalen and van Leeuwen 2002), see example in Fig. 1. Actually, the PNEC-based Dutch Environmental Quality Criteria (EQC) are estimated in this way (de Bruijn et al. 1999) as well as the PNEC in the USES-LCA method (Huijbregts 2001), when sufficient data are available, i.e. at least 4 NOEC values according to de Bruijn et al. (1999).

Within LCIA, a substance's characterisation factor (CF) for ecotoxicity can be expressed as the change in the ecotoxicity effect indicator (EEI) due to a change in the environmental concentration (dC) for every kilogram emitted of the substance. When the effect indicator is derived from an assessment factor-based PNEC approach, the expression becomes:

$$CF = EEI \cdot dC = \frac{1}{PNEC} \cdot dC \,, \quad (1)$$

And, if PNEC is SSD based (i.e. PNEC = $HC5_{NOEC}$), then

$$CF = EEI \cdot dC = \frac{1}{HC5_{NOEC}} \cdot dC \quad (2)$$

2 PAF-Based Impact Approaches

The Potentially Affected Fraction of species (PAF) may be described as the fraction of species in a (generic) ecosystem/community that is expected to be (potentially) affected above its no-effect level or a predefined effect level at a given environmental concentration of a toxicant (adopted from Traas et al. 2002).

The PAF approaches are based on the principles of Species Sensitivity Distribution (SSD) which is a statistical distribution describing the variation among a set of species in toxicity of a certain substance or mixture, see Fig. 1.

The overall SSD approach or PAF approach is used or proposed to be used in the following identified ways in LCIA:

- Multi substance PAF (msPAF) or combi-PAF approach
- 'Marginal PAF increase' approach (tangential or marginal gradient)
- 'Average PAF increase' approach, HC5 (secantial or average gradient)
- 'Average PAF increase' approach, HC50 (secantial or average gradient)

The different PAF approaches are described shortly in Section 2.1 and in more detail in Appendix A (see Online Edition).

2.1 'Marginal PAF increase' approach

The multi substance PAF (msPAF) method is used within LCIA as part of the 'marginal PAF increase' approach in the Eco-indicator 99 method (Goedkoop and Spriensma 2001a, 2001b), where it is referred to as 'combi-PAF'.

The msPAF method addresses the simultaneous exposure to several different substances. The first step is to calculate a single substance PAF curve (like in Fig. 1) for each substance and to estimate the geometric mean (GM) of the NOEC values (we assume that it's a NOEC-based SSD as in Eco-indicator 99), see Section A1 in Appendix A for details (see Online Edition). For each substance, all NOEC values are then divided by the GM for the substance. Hereby, the NOEC values are normalized to Hazard Units (HU). These HUs together with their corresponding PAF values are used to construct the msPAF curve, see Fig. A3 in Appendix A for an example (see Online Edition). By scaling each concentration to HU, the msPAF method makes it possible to estimate the potentially affected fraction of species due to a mixture exposure. If all the toxicants in the mixture have the same Toxic Mode of Action (TMoA), e.g. narcotic, we can use the principles of Concentration Addition (CA) and just add the HU for each toxicant, but if different TMoA are involved we have to use the more complicated procedure for Response Addition (RA), see Appendix A, Section A2 (see Online Edition). Huijbregts et al. (2002) and Pennington et al. (2004) have described how to use RA in connection with the 'marginal PAF increase' approach and the 'average PAF increase' approach, respectively.

In the 'marginal PAF increase' approach, the EEI is expressed as the marginal increase in the msPAF of the community/ecosystem SSD in question, due to exposure to an emitted toxicant. The aim is to express the potential impact from an emitted toxicant by the marginal increase in the number of species having their NOEC values exceeded taking the background impact (i.e. the number of species already having their NOEC exceeded) into account. Since the estimated average background impact level in The Netherlands is 22% (i.e. 22% of the species have their NOEC exceeded), the point on the msPAF-curve corresponding to msPAF = 0.22 is chosen as the working point in the Eco-indicator 99 method (Goedkoop and Spriensma 2001a, 2001b). When calculating the marginal increase in msPAF (i.e. dmsPAF) due to a marginal increase in the environmental concentration, the slope (i.e. slope factor γ) of the tangent to the working point is used. At the working point msPAF = 22%, this slope factor is determined as 0.59. This gives the following general equation:

$$CF = dmsPAF = EEI \cdot dC = \frac{\gamma}{HC50_{NOEC}} \cdot dC \quad (3)$$

If γ is equal to 0.59, then:

$$CF = \frac{0.59}{HC50_{NOEC}} \cdot dC \quad (4)$$

$HC50_{NOEC}$ is in this case the geometric mean (GM) of the single substance SSD values (i.e. NOECs) of the substance in question.

The reason for dividing by HC50 in Eq. 3 and 4, and the following equations on CF, is that the increase in concentration (dC) must be expressed in HUs on which the slope is defined, see Fig. A3 in Appendix A (see Online Edition).

2.2 'Average PAF increase' approaches

2.2.1 HC5-based approach

Instead of using a tangential gradient, as in the 'marginal PAF increase' approach, Pennington et al. (2004) argue for using a secant gradient ('average PAF increase' approach), especially at low exposure concentrations (below HC5) where the shape of the PAF curve becomes very uncertain and dependent on the distribution model chosen. The secant or average gradient is a linear gradient between the origin (of the PAF curve) and a chosen working point on the curve corresponding to PAF = 0.05, for example, assuming that the background impact level is below PAF = 0.05. The general characterisation factor equation for this approach is:

$$CF = dPAF = EEI \cdot dC = \frac{0.05}{(HC5/HC50)/HC50} \cdot dC \quad (5)$$

As shown by Pennington et al. (2004), the factor (HC50/HC5) may be expressed as $10^{2.94\beta}$ (where β is the scale parameter), thus leading to the following general equation:

$$CF = \frac{0.05 \cdot 10^{2.94\cdot\beta}}{HC50} \cdot dC \quad (6)$$

2.2.2 $HC50_{EC50}$-based approach

As recently proposed as a best available practice in LCA by Pennington et al. (2004), and as implemented in the method for Assessment of the Mean Impact on ecosystems, AMI (Payet and Jolliet 2005, Payet 2004, 2005), PAF = 0.5 may be used as a working point for the average gradient (see example in Fig. A3 in Appendix A, Online Edition). In the AMI method, two statistical estimators have been explored for the assessment of the average toxicity of chemicals. A non-parametric estimator, using the median as the $HC50_{EC50}$ value combined with bootstrap statistics for the estimate of its uncertainty (Payet and Jolliet 2005), and an estimator based on the assumption of lognormal distribution of data using the GM as $HC50_{EC50}$ and the student's t-statistics for its confidence interval (Payet 2004, 2005). In both cases, the PAF curve is based on EC_{50} values for chronic toxicity (instead of NOEC values as in the Eco-indicator 99 method, for example). Background impact level is assumed not to be relevant in probably almost all cases. For the 'average PAF increase' approach based on $HC50_{EC50}$, the general characterisation factor equation is:

$$CF = dPAF = EEI \cdot dC = \frac{0.5}{HC50_{EC50}} \cdot dC \quad (7)$$

3 Damage Approaches

The description below, of the three damage approaches, is based on Payet and Larsen (2006).

3.1 Recovery time approach

One of the damage approaches, the 'recovery time approach' used as 'media recovery' is actually used in the Eco-indicator 99 method (Goedkoop and Spriensma 2001a, 2001b) and the IMPACT 2002+ method (using AMI) (Jolliet et al. 2003) when estimating the potentially vanished fraction of species (PVF).

The media recovery model is based on the assumption that the media quality (here defined as the level of toxicants in the media) is linked directly to the biodiversity. Species are considered as having disappeared as soon as the toxicant concentration in the ecosystem reaches a certain level, and considered as having reappeared when the toxicant disappears, e.g. by degradation. This model is thus directly linked to the fate modelling. This is the easiest way to link the PAF and the PVF and it is based on two rough assumptions:

- The assumption of the equivalence time between disappearance and recolonisation of species. This requires at least some knowledge about the life history of species, which is not available for most of the species present in the environment.
- The assumption of the equivalent diversity before and after the toxic impact. This assumption would not be valid for a large scale assessment, where the reduction of populations would lead to an important genetic drift, and, therefore, a reduction of genetic diversity.

These working assumptions have not been tested yet, and the models are currently at a research level.

Nevertheless, these models may be linked to the PAF-based impact approaches because PVF may be expressed as a function of PAF depending on the toxicant concentration, on the one hand, and the time of exposure on the other hand. Both the Eco-indicator 99 method (Goedkoop and Spriensma 2001a, 2001b) and the AMI method (Jolliet et al. 2003) express the results of ecotoxicological impact in PAF units. In order to translate the result into an endpoint indicator, Eco-indicator 99 assumes a factor 10 of increase in concentration of exposure to express the result in PVF instead of PAF. This factor of 10 is included since NOEC values are used for building the msPAF curve, implicitly assuming that an exposure 10 times higher than NOEC will lead to the extinction of the species. The AMI method uses the same basis, but considers the fraction of affected species based on chronic EC_{50} values instead of the NOECs, and therefore a factor 2 instead of 10 is applied for translating the PAF to PVF, thus inherently assuming that half of the species exposed above their EC_{50} level will become extinct. However, the factor is disputable since other research results shows that a 50% effect, for example on reproduction, will always result in extinction of the population after a hundred years of exposure (Snell and Serra 2000), meaning, in this perspective, the that factor would be 1 and PVF=PAF.

3.2 Time to extinction approach

The environmental threats should not cause immediate extinction of a population, but will shorten the expected time to extinction (Hakoyama and Iwasa 2000). Based on stochastic population models, some models, like the Mean Extinction Time (MET), have been developed to quantify the expected survival of species exposed to a habitat size reduction or an environmental pollution (Lande 1998). Among the consequences of a toxicant effect is a reduction in the growth rate of the population. The estimation of this decrease can be translated into a MET risk, corresponding to the decrement of the intrinsic growth rate which can be assessed with a mathematical model (Tanaka and Nakanishi 2000). These models may include both the normal environmental fluctuation in population growth rate and the random catastrophes. The required parameters for these sorts of models are generally:

- Reproduction rate, based on the life history of the organisms
- Carrying capacity of the media, which is used to assess the initial population size

Only one attempt has yet been performed to include these models in LCIA (Itsubo et al. 2003, Itsubo and Inaba 2003, Narita et al. 2004).

3.3 Genetic diversity

Instead of using biodiversity as a basis for the endpoint/damage modelling, use of genetic diversity could be a good alternative in solving some of the problems with diversity within species contra diversity between species, and the problem with vulnerability of species after repeated exposure to toxicants. Facilitating the interpretation of the changes in genetic diversity, a model has recently been presented linking the phenotypic diversity and the ecosystems' functioning. This model could provide a way to translate the changes in genetic diversity (which is directly linked with the phenotypical variance reduction) in terms of modification of an ecosystems' functioning (Norberg et al. 2001). The strength of the study is to focus on the functional group of species as the basic unit of ecosystems, looking at its sensitivity to the changes in diversity. Multispecies models are reduced to three equations that represent the total biomass in the community, the average phenotype, and the phenotypic variance. This model has the strengths of linking the evolutionary dynamic to the ecosystem functioning (Tilman 2001), and of expressing the results in terms of quantitative change in biomass production in the ecosystems. Such a model is promising, but the data are not available to support the computation of the reduction in biomass production from the average phenotype and the reduction in phenotypic variance resulting from toxic pressure.

4 Assessment Criteria

The different approaches are evaluated against the following criteria, which are partly based on Hauschild and Pennington (2002):

Compatibility. This criterion deals with the degree to which the method/approach, including its assumptions and interpretation, is compatible with the methodological requirements of LCA. The main requirements include the fitness for comparison, i.e. use of best estimate (non-conservative) and additivity of the indicator.

Environmental relevance. The indicator can be more or less relevant for the effect we want to indicate, i.e. effects on (an) ecosystem(s), here interpreted as potential impact or damage on 'ecosystem health' represented by species diversity as described in the Introduction. As an example, an indicator only taking one acute value (e.g. LC50) into account is less environmentally relevant than an indicator taking chronic data on several taxa from three trophic levels into account. It should be possible to interpret the result in terms of either impact (midpoint) or damage (endpoint) depending on how far the indicator is modelled along the environmental mechanism. As environmental relevance is abstract and elusive to define, it might seem irrelevant to include this criterion. Due to the inherent characteristics of LCA (focus on functional unit, not full output from processes, aggregation over time and space, lack of knowledge about background situation in receiving environment), LCIA can never predict actual effects and is hence not possible to validate in a strict sense against measurements. Anyway, we want the results of an LCIA to be in accordance with (or at least related to) our experience of real effects occurring in the environment. So, even though the connection is weak in practice, being aware of the 'environmental relevance' is very important for LCIA. Furthermore, if we chose a very simple method based on only one parameter (e.g. molecular weight), it may score high in all criteria used here except the one on 'environmental relevance'.

Reproducibility. To what extent will the method, its description and interpretation allow different practitioners to come up with the same substance-specific indicators, considering also the variability of the input data? As an example PNEC based approaches using one (i.e. the lowest) NOEC value are very sensitive to the database used, whereas average approaches are less sensitive to changes in database and therefore more reproducible.

Data demand. The number and kind of data needed for calculating the indicator is crucial for how easy it actually is to produce indicators (e.g. data which is difficult to access will demand more time) and the number of chemicals for which the calculation is possible (no relevant data, no indicator). PNEC approaches, for example, only demanding one acute value, have a substantially lower data demand than the 'marginal PAF increase' approach with its requirement of at least 4 NOEC values (or much more if a background level is to be estimated).

Data availability. If the data needed for the method has a low availability, only a limited number of indicators can actually be calculated. This criterion is assessed on the basis of the availability of the required types of data in the databases (e.g. AQUIRE) and handbooks available for practitioners. Availability of this kind of data is described in Posthuma et al. (2002), for example.

Quantification of uncertainty. Is it possible to calculate/estimate the uncertainty of the indicators calculated by the method, knowing that the uncertainty will give an indication of the reliability of the result?

Transparency. For the method to be generally accepted and understood by the user, it is important that it is transparent. It should not be too complex, but explainable to the practitioner. It should be well-documented, and a manual calculation should be possible. So, the main issue here is how easy it is to understand and use the method. More simple methods based on PNEC are easier to understand and use than more complex ones based on PAF.

Spatial differentiation. To what degree is it possible to include spatial differentiation for a given approach? An approach like the one based on the 'marginal PAF increase' is designed for spatial differentiation (including background). The 'average PAF increase' approach may also be used (if β is not fixed), but the PNEC approach is not especially developed for this purpose in the context of LCIA.

5 Results

The result of the evaluation of the PNEC- and PAF-based approaches against these criteria is shown in **Table 1.** The plusses in the table can only be compared within each criterion and not between criteria and are only a relative indication of the evaluation which is further specified in Section 6. Due to the development stage of the damage approaches, these are not evaluated in a systematic way as in Table 1, but only commented upon in Section 6.3.

6 Discussion

6.1 AF-based PNEC approaches

A general problem with the AF-based PNEC approaches (see Eq. 1) in LCIA is their use of conservative estimates. Therefore, only between one and two plusses (+–++) is allocated to the AF-based PNEC approaches on compatibility in Table 1. Furthermore, the PNEC approach is based on the lowest ecotoxicity value and, therefore, relatively sensitive to the database used if not all available data (i.e. all data sources) are included.

The main problem when using an AF-based PNEC approach based only on acute data is that it has a low environmental relevance (+). Besides local situations with high exposure, the only kind of effects that we can expect to occur in the 'real' ecosystems are chronic. LCA typically deals with chronic exposure while the available ecotoxicity data mostly involve acute data. If we try to solve this problem by extrapolating acute data to chronic data (typically by applying a factor of 10), we face a problem with different kinds of TMoA within and between different kinds of chemicals and interspecies differences, thus resulting in different acute to chronic ratios (ACRs). These may be up to a factor 500 if based on acute and chronic average toxicity over species (de Zwart 2002), and, if based on single species, in the range of 0.79–5,500 (Forbes and Calow 2002). The inclusion of chronic data may therefore improve the environmental relevance significantly. Reproducibility is fair (++) because of relatively high general availability of acute data. Concerning transparency, low data demand and high data availability, the score is very high (++++) because this approach based only on acute data is simple and needs only one acute data value for which the availability is relatively high. On both spatial differentiation and quantification of uncertainty, the score is low (+) because these issues are neither implemented nor readily possible.

The AF-based PNEC approach based solely on chronic data to some degree solves the problem with lack of environmental relevance (+++) of the approach based only on acute data. In this case, however, we run into the problem that the number of chemicals with sufficient chronic data is very limited (score on data availability '++'). Only for 1000 to 1500 chemicals are chronic data available, whereas acute data are available for 6000 to 10000 chemicals (rough estimates based

Table 1: Assessment of PNEC- and PAF-based approaches

Criteria	PNEC-based approaches				PAF-based approaches		
	AF-based PNEC (only acute data)	AF-based PNEC (only chronic data)	AF-based PNEC chronic (preferred) and acute data)	SSD-based PNEC ($HC5_{NOEC}$)	Marginal PAF increase' (fixed β value)	Average PAF increase, HC5 (fixed β value)	Average PAF increase, HC50
Compatibility	+	++	+(+)	++	++	++	+++
Environmental relevance	+	+++	++(+)	+++	+++	+++	+++
Reproducibility	++	+(+)	+(+)	++(+)	+++	++(+)	+++
Transparency	++++	+++(+)	+++(+)	++	+(+)	+(+)	+(+)
Low data demand	++++	+++	+++(+)	++	+	++	++
High data availability	++++	++	++++	+(+)	+	+(+)	+(+)
Spatial differentiation possible	+	+(+)	+	++	+++	++ [a]	+(+)
Quantification of uncertainty included	+	+	+	++ [c]	++ [c]	++ [c]	++++ [b]

++++: Very high degree of fulfilment; +++: High degree of fulfilment; ++: Moderate degree of fulfilment; +: Low degree of fulfilment

[a]: β not fixed; [b]: Implemented in AMI (Payet 2004, 2005); [c]: Not implemented but possible

on de Zwart (2002), Allanou et al. (1999), data availability in ECOTOX (2002), Payet 2004, 2005). If EU's REACH system for registration and reporting of data on chemicals (EC 2003b) is implemented in the coming years, the number of chemicals for which ecotoxicity data become available will probably increase significantly, but primarily due to a higher number of acute data. The AF-based PNEC approach based solely on chronic data is a bit less transparent (+++(+)) than the one based solely on acute data. It has a data demand (+++) of 2–3 NOEC values. Spatial differentiation (+(+)) may be a bit more realistic than if based solely on acute data, but quantification of uncertainty (+) is neither implemented nor readily possible.

By combining the two above-mentioned approaches in an AF-based PNEC approach where chronic data is preferred (the way it is typically done today), we still have problems related to extrapolation from acute to chronic and environmental relevance (++(+)) in the cases were we are forced to use acute data. In addition, we acquire a problem with biased ecotoxicity indicators because some are based on acute data (far from the PNEC endpoint and with high uncertainty) and others on chronic data (close to the endpoint and with less uncertainty). This is especially a problem if conservative extrapolation factors are used, and this is typically the case today. Reproducibility (+(+)) and transparency (+++(+)) is the same as for the approach based solely on acute data. The data demand is relatively low (+++(+)) as this approach is also able to work on only one acute data value and therefore the data availability is high (++++). For spatial differentiation (+) and quantification of uncertainty (+), the situation is identical to the approach based solely on acute data.

The SSD-based PNEC approach (see Eq. 2) has the potential of being more environmentally relevant (+++) than the AF-based approaches because all available chronic data are exploited instead of a single one (the lowest), as in the AF-based approaches. But if the data used do not represent the ecosystem we want to relate our estimates to (e.g. when the selection of ecotoxicity is without relevance for the trophic structure of the ecosystem) and/or if assessment factors are used anyway to estimate chronic values from acute ones, this approach is most probably not more environmentally relevant than the AF-based PNEC approaches. Another problem with the SSD-based PNEC approach, as compared to the AF-based approaches, is that the data demand (++) is higher on chronic data where the data availability (+(+)) is low. At least 4 NOEC values (de Bruijn et al. 1999) or more than 10 NOEC values to have a stable result (van Straalen and van Leeuwen 2002) are needed to derive $HC5_{NOEC}$, as opposed to 2–3 NOEC values for obtaining the lowest AF in the AF approaches including chronic data (e.g. Hauschild et al. 1998, EC 2003a). The higher data demand for $HC5_{NOEC}$ is reflected in the generic risk assessment recommendations in the EU TGD (EC 2003a), stating that if $HC5_{NOEC}$ is used for estimating PNEC, at least 10 NOEC values are needed covering at least 8 different taxonomic groups. The estimation procedure of the SSD-based PNEC approach is less transparent (++) than that of the AF-based approach, but a quantification of uncertainty (++) can be performed if it is tested or assumed that the data fits the distribution function used. Reproducibility is assessed to be higher (++(+)) than for the other PNEC approaches because at least four data values are included in the estimation of PNEC and not just one, i.e. the lowest. Anyway, the uncertainty for estimating HC5 is relatively high (see Fig. A4 in Appendix A, for example, Online Edition) and sensitive to the inclusion of data from test on insensitive species (lowering the HC5 value). The possibilities for spatial differentiation (++) is higher for this approach than for the other PNEC approaches due to the inclusion of data values from several species.

6.2 PAF-based approaches

All the PAF-based approaches are making use of the SSD framework and are therefore associated with its advantages and drawbacks. Two general problems are mentioned above i.e. the haphazard use of ecotoxicity data disregarding their representativity for the trophic structure of an ecosystem and the high demand on data of low availability. Another problem is that for 25%–50% of the chemicals with available data, the SSD does not follow the parametric distribution functions typically used, i.e. lognormal (Newman et al. 2002, Payet 2004, 2005). The non-parametric version of the AMI method (Payet and Jolliet 2005) avoids the last mentioned problem by using a distribution-free, non-parametric SSD method making use of the median and bootstrap technique for calculating confidence limits. Advantages of the PAF approaches are that modelling the effects of mixtures is possible, and that modelling to endpoint (damage) seems at least theoretically possible. If spatial differentiation is considered, the inclusion of the species sensitivity may also be of some advantage as compared to the AF-based PNEC approach. The 'marginal PAF increase' approach (see Eq. 3) seems most powerful in spatial differentiation because of the possibility of defining the spatially differentiated background level by choosing different working points on the msPAF curve, see Fig. A3 in Appendix A (Online Edition). On compatibility with the methodological framework of LCA, the PAF–based approaches have a possibility of being non-conservative in providing a best estimate, and they are therefore given the score '++' except for the average approach based on HC50, which is given the score '+++' due to its use of effect data instead of no-effect data. The PAF-based approaches all exploit the whole set of chronic data with the possibility of having a relatively high environmental relevance (+++). Transparency (+(+)) of all PAF-based approaches is lower than for the AF-based types due to a more complicated estimation principle.

Because the 'marginal PAF increase' approach includes the background impact level, it has a huge data demand on background concentration of toxicants and background PAFs (to define the working point) in order to create the multi substance PAF curve (i.e. determine the β value), if it is going to cover more than just the Netherlands. When the msPAF curve is created, the data demand is reduced to at least 4 NOEC values for each chemical (Goedkoop and Spriensma 2001a, 2001b), and the score for both data demand and data availability is '+'. This data demand of 4 NOEC values for each chemical is met for a maximum of 200 chemicals (de Zwart 2002). In the Eco-indicator 99 method making use of this

approach, it has only been possible to calculate ecotoxicity effect indicators for about 40 substances (Goedkoop and Spriensma 2001a, 2001b). However, the fact that this approach includes background impact and that it is at least in theory possible to estimate the impact level for different places (countries, habitats, e.g. forests, agricultural land, etc.), makes this approach more relevant for spatial differentiation (+++) than the others. As the indicator of this approach is based on the GM of 4 NOECs, it is assessed to have a high reproducibility (+++). Quantification of uncertainty is not yet implemented but possible (+(+)).

The 'average PAF increase' approach with working point at HC5 (see Eq. 5) has a lower data demand than the 'marginal PAF increase' approach for the creation of the msPAF curve, because it does not include background impact (i.e. it is assumed that the background impact is below PAF=0.05). For estimation of the ecotoxicity effect indicator on each chemical, the data demand is the same as for the marginal approach, if we assume that a fixed β value is used and that the msPAF curve is NOEC-based. The fact that the β value is dependent on the TMoA may cause more than a factor 100 error in the estimated effect indicator in both the 'marginal PAF increase' approach (Huijbregts et al. 2002) and the 'average PAF increase' approach based on $HC5_{NOEC}$ (Pennington et al. 2004), if a fixed β value is used. If it is not accepted to use a fixed β value, and an exact one is not known, the $HC5_{NOEC}$ value has to be determined (see Eq. 5) in the 'average PAF increase' approach based on $HC5_{NOEC}$. In this case, the data demand is what is needed to determine the $HC5_{NOEC}$, i.e. at least 10 NOECs to have a stable result as described above. The reproducibility (++(+)), the data demand (++), data availability (+(+)) and possibilities for quantification of uncertainty (++) are assessed to be at the same level as for the SSD-based PNEC approach with possibilities for spatial differentiation (++) by using a variable β value.

The 'average PAF increase' approach based on $HC50_{EC50}$ (see Eq. 7) is not dependent on a β value because it is assumed that the increase in PAF, due to an increase in the concentration of the toxicant, can be described by an average linear gradient starting from (1, 0.5) and having a slope of 0.5 on a standardised PAF curve showing the concentration of the toxicant on the X-axis in hazard units (HUs), see Fig. A3 in Appendix A (Online Edition). As compared to an average gradient based on HC5, taking variation on β values into account, the uncertainty of assuming an average gradient based on HC50 is below a factor 10 for most chemicals, as shown by Pennington et al. (2004). This estimation is valid for an interval of 0.2–0.7 for acute β values covering the main part of the 89 chemicals tested by de Zwart (2002). However, at least 17 of the tested chemicals have acute β values above 0.7. Furthermore, the range of the chronic β values is higher, with at least 19 chemicals having chronic β values above 0.7 and at least 7 having chronic β values below 0.2. Actually, the observed range in chronic β values by de Zwart (2002) is 0.02–1.65, but extreme values (at least above 1.25) are to be considered as artefacts due to a very poor data foundation, i.e. typically only 3–10 species tested, and not the 25–50 needed to reach a stabilized β value (de Zwart 2002).

The $HC50_{EC50}$-based 'average PAF increase' approach is implemented in the AMI method (Payet and Jolliet 2005, Payet 2004, 2005). In this case, chronic EC_{50} values are used instead of chronic NOEC values as typically used in PAF approaches. In this way, a general problem with NOEC values is avoided, i.e. the problem that a NOEC value is determined as the highest measured value which is not statistically different from a control value in a laboratory test. The determined NOEC value is therefore dependent on the test design used (the concentrations that were actually tested), and as a consequence not necessarily just below the lowest concentration where the chronic effect actually occurs for the organism tested. Contrary to all the other EEI approaches, the AMI method includes quantification of uncertainty (++++). The non-parametric version of the AMI method (Payet and Jolliet 2005), using the median for estimating $HC50_{EC50}$, requires a minimum of 5 chronic EC_{50}s for the bootstrap technique to estimate confidence limits. Procedures for estimating the median-based $HC50_{EC50}$ and the confidence limits on only three acute data (representing three taxonomic groups) are described, but based on assessment factors (Payet et al. 2002). As an alternative to the non-parametric version, the parametric version of AMI can be applied with a minimum of three different EC50s from 3 different taxonomic groups (Payet 2004, 2005), assuming log-normal distribution of data, and using the geometric mean for estimating $HC50_{EC50}$. Both acute and chronic data may be used. The AMI method suffers from the general low availability of chronic data, and the use of assessment factors (acute to chronic) is needed for high substance coverage. Furthermore, confidence limits based on only three data values typically become quite wide, making the statistically differentiation between substances impossible. The fact that most chronic ecotoxicity data are found as NOEC values creates the need for a 'new' kind of assessment factors to be used when estimating chronic EC_{50} values from NOEC values. The AMI method's score on data demand is therefore assessed to '++' and on data availability to '+(+)'. As β is fixed, the possibilities for spatial differentiation is assessed to '+(+)'. Finally, it should be noticed that the use of the geometric mean of the EC50s improves the stability of the indicator regarding the species tested and makes it less sensitive to differences in the actual ecotoxicity data used (e.g. data from different databases). Hereby, the reproducibility (+++) is improved.

6.3 Damage-based approaches

With the exception of the 'media recovery' approach, which depends directly on the fate model, all three approaches for damage modelling have a high environmental relevance but very low data availability. They are all at a very early research stage, especially the approach on genetic diversity. Despite this fact, the recovery time approach used as media recovery has been used in some attempts to include damage modelling in the Eco-indicator 99 method (Goedkoop and Spriensma 2001a, 2001b), and most recently in IMPACT 2002+ (AMI) (Jolliet et al. 2003). The damage approaches are further evaluated in Payet and Larsen (2006).

7 Conclusions and Recommendations

Based on the evaluation, none of the existing methods can be recommended as optimal in their present form. Different directions for improvement and further development of existing approaches, and development of new ones, exist as well. There seems to be at least three main directions to go:

- Improving the assessment factor-based PNEC approaches making them less risk assessment-oriented and more suitable for LCIA. The goal involves development of non-conservative assessment factors including uncertainty estimates (confidence limits), and, if possible, taking toxic mode of action into account. However, the problem with instability of the indicator due to its dependence on the choice of the database still remains.
- Improving the chemical coverage and the environmental relevance of the 'PAF related' approaches. The main problem here seems to be the lack of data and the fact that the way these approaches have been used till now does not reflect effects on the ecosystem in a more accurate way than the assessment factor-based PNEC approaches. The goal will be to make procedures for a more environmentally relevant application (e.g. more realistic, not haphazard representation of species on each trophic level). There is also a need to improve the chemical coverage by fitting the approach to a low data availability, and further to utilize, improve and develop the inclusion of mixtures and damage modelling.
- Further development of the 'media recovery' damage approach or development of new damage-based indicator based on the 'time to extinction' approach or 'changes in genetic diversity'.

For damage modelling, the 'media recovery' approach seems to be the most realistic way to go if a method is to be functional for LCIA within the near future. The media recovery approach can be coupled with the PAF approaches, but, for example, needs further development on the connection between media recovery and recovery/recolonization of species populations. Taking into account that the two other damage approaches are at an even earlier developmental stage than the 'media recovery' model, and that the availability of the needed data is very poor, it is probably not realistic to attain practical; useable methods based on these approaches in the near future. However, from a theoretical point of view, the approaches based on mean extinction time and genetic diversity are very attractive.

For the assessment factor-based PNEC approaches; the main problem is that they are founded in the first tier of a multi-tiered risk assessment and therefore conservative, which is not compatible with the comparative framework of LCIA. A way to deal with this problem could be to try to develop non-conservative assessment factors taking the huge work on acute to chronic ratios already done (e.g. Chapman et al.1998, Solbe et al. 1998, Länge et al.1998, Forbes and Calow 2002) as a starting point, and maybe trying to differentiate the assessment factors by TMoA. However, as the PNEC approach is no-effect based (i.e. NOEC based), it will still suffer from the uncertainty of measured NOEC values due to variation in test design, and the potential dependence of the lowest toxicity value on the choice of database to characterize the toxicity of the substance.

If we accept using a fixed β value in Eq. 6, which is the most appropriate if the method is going to be functional in a normal LCIA context, then, despite the described differences in theoretical foundation, all PAF approaches described here lead to the following general characterisation factor equation:

$$CF = dPAF = EEI \cdot dC = \frac{k}{HC50} \cdot dC \quad (8)$$

The constant k in Eq. 8 may be 0.59, as in the Eco-indicator 99 method (Goedkoop and Spriensma 2001a, 2001b), 0.5, as in the average HC50-based approach or 0.2–43, as in the average HC5-based approach (Pennington et al. 2004). So, in the comparative approach applied in LCIA, there is no difference in practise between the PAF approaches, as long as the same value for HC50 and the same value for change in concentration (dC) are used. The key element in the effect indicator part (k/HC50) of Eq. 8 therefore becomes HC50.

The crucial point in the determination of a PAF-based ecotoxicity effect indicator is thus the data used and the principles applied for determining the HC50 value of each toxicant. As mentioned above, the HC50 may be estimated by use of e.g. NOEC values or EC_{50} values and based on the non-parametric median or the parametric geometric mean. Furthermore, the actual data used may, for example, reflect haphazard representation of species on each trophic level or a more realistic and consciously chosen representation of the structure of the ecosystem/community in question. The choice of data and estimation principle may therefore have significant influence on the outcome, especially when the amount of available data on each toxicant is low, which is the typical case within LCIA handling many chemical emissions. These issues are addressed in a second paper by the authors Larsen and Hauschild (2006).

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References

Aldenberg T, Jaworska J, Traas TP (2002): Normal Species Sensitivity Distributions and Probabilistic Ecological Risk Assessment. Chapter 5. In: Species Sensitivity Distribution in Ecotoxicology. Posthuma L, Suter II GW, Traas TP (eds), Lewis, Boca Raton, FL, USA, pp 49–102

Allanou R, Hansen BG, van der Bilt Y (1999): Public Availability of Data on EU High Production Volume Chemicals, <http://ecb.jrc.it/>

Bruijn JCT de, van Leeuwen K, van der Plassche E, Sijm D, van der Weiden M (1999): Environmental Risk Limits in The Netherlands. Part I. Procedures In: Könemann H (ed), Environmental Risk Limits in The Netherlands. Report No. 601640 001. RIVM, Bilthoven, The Netherlands, pp 1–33

Calow P (1998): General Principles and Overview. Chapter 1. In: Calow P (ed), Handbook of Ecotoxicology. Blackwell Science, Oxford, Great Britain, pp 1–5

Campbell RC (1974): Statistics for biologists. Second edition. Cambridge University Press, Cambridge, Great Britain

Chapman PM, Fairbrother A, Brown D (1998): A critical evaluation of safety (uncertainty) factors for ecological risk assessment. Environ Toxicol Chem 17 (1) 99–108

EC (2003a): Environmental Risk Assessment. Chapter 3 (Part II) In: Technical Guidance Document on Risk Assessment in support of Commission Directive 93/67/EEC on Risk Assessment for New Notified Substances, Commission Regulation (EC) No. 1488/94 on Risk Assessment for Existing Substances and Directive 98/8/EC of the European Parliament and the Council

concerning the placing of biocidal products on the market. Institute for Health and Consumer Protection. European Commission. ECB, JRC, Ispra, Italy
EC (2003b): Proposal for a Regulation of the European Parliament and of the Council concerning the Registration, Authorisation and Restriction of Chemicals (REACH), establishing a European Chemicals Agency and amending Directive 1999/45/EC and Regulation (EC) {on Persistent Organic Pollutants}. COM (2003) 644, Brussels, 29 October 2003
ECOTOX (2002): ECOTOXicology Database system. US EPA: <http://www.epa.gov/ecotox/>
Forbes VE, Calow P (2002): Extrapolating in Ecological Risk Assessment: Balancing Pragmatism and Precaution in Chemical Controls Legislation. BioScience 52 (3) 249–257
Goedkoop M, Spriensma R (2001a): The Eco-indicator 99. A damage oriented method for Life Cycle Impact Assessment. Methodology Report. Third edition. Pré Consultants, Amersfoort, The Netherlands <http://www.pre.nl>
Goedkoop M, Spriensma R (2001b): The Eco-indicator 99. A damage oriented method for Life Cycle Impact Assessment. Methodology Annex. Third edition. Pré Consultants, Amersfoort, The Netherlands, <http://www.pre.nl>
Hakoyama H, Iwasa Y (2000): Extinction risk of a density-dependent population estimated from a time series of population size. J Theor Biol 204, 337–359
Hauschild M, Pennington D (2002): Indicators for Ecotoxicity in Life-Cycle Impact Assessment. Chapter 6 In: Udo de Haes HA, Finnveden G, Goedkoop M, Hauschild M, Hertwich E, Hofstetter P, Jolliet O, Klöpffer W, Krewitt W, Lindeijer E, Müller-Wenk R, Olsen SI, Pennington D, Potting J, Steen B (eds), Life-cycle Impact Assessment: Striving Towards Best Practice. SETAC-Europe, Brussels, Belgium, pp 149–176
Hauschild M, Wenzel H, Damborg A, Tørsløv J (1998): Ecotoxicity as a criterion in the environmental assessment of products. Chapter 6. In: Hauschild M, Wenzel H (eds), Environmental Assessment of Products: Volume 2: Scientific background. London, Chapman & Hall, London, Great Britain, pp 203–314
Huijbregts M (2001): Uncertainty and variability in environmental life-cycle assessment. Ph.D. Thesis University of Amsterdam. ISBN 90-76894-05-1
Huijbregts M, Thissen U, Guinée JB, Jager T, Kalf D, Van de Meent D, Ragas AMJ, Wegener Sleeswijk A, Reijnders L (2000): Priority assessment of toxic substances in life cycle assessment. Part I: Calculation of toxicity potentials for 181 substances with the nested multi-media fate, exposure and effects model USES-LCA. Chemosphere 41, 541–573
Huijbregts M, Van de Meent D, Goedkoop M, Spriensma R (2002): Ecotoxicological Impacts in Life Cycle Assessment. Chapter 20 In: Posthuma L, Suter II G W, Traas T P (eds), Species Sensitivity Distributions in Ecotoxicology. Lewis, Boca Raton, FL, USA, pp 421–433
ISO (2000): ISO 14042: Environmental management – Life cycle assessment – Life cycle impact assessment. International Organisation for Standardisation, Geneva, Switzerland
Itsubo N, Ii R, Abe K, Nakagawa A, Hayashi K, Inaba A (2003): Biodiversity Damage Assessment – Applying the Theory of Biology in LCIA. Slides from platform presentation (WE2/8) at the SETAC Europe 13th Annual Meeting at Hamburg, Germany, April 27–May 1, 2003
Itsubo N, Inaba A (2003): A New LCA Method: LIME has been completed. Int J LCA 8 (5) 305
Jolliet O, Margni M, Charles R, Humbert S, Payet J, Rebitzer G, Rosenbaum R (2003): Impact 2002+: A New Life Cycle Impact Assessment Methodology. Int J LCA 8 (6) 324–330
Lande R (1998): Anthropogenic, ecological and genetic factors in extinction and conservation. Researches on population ecology 40 (3) 259–269
Larsen HF, Birkved M, Hauschild MZ, Pennington DW, Guinée JB (2004): Evaluation of selection methods for toxicological impacts in LCA: Recommendations for OMNIITOX. Int J LCA 9 (5) 307–319
Larsen HF, Hauschild M (2006): GM-troph – A low data demand ecotoxicity effect indicator for use in LCIA. Int J LCA, OnlineFirst <DOI: http://dx.doi.org/10.1065/lca2006.12.288>
Länge R, Hutchinson T, Scholz N, Solbe J (1998): Analysis of the ECETOC Aquatic Toxicity (EAT) database. II – Comparison of acute to chronic ratios for various aquatic organisms and chemical substances. Chemosphere 36 (1) 115–127
Molander S, Lidholm P, Schowanek D, Recasens M, Fullana P, Christensen FM, Guinée JB, Hauschild MZ, Jolliet O, Carlson R, Pennington DW, Bachmann TM (2004): OMNIITOX – Operational life-cycle impact assessment models and information tools for practitioners. Int J LCA 9 (5) 282–288
Narita N, Nakahara Y, Morimoto M, Aoki R, Suda S (2004): Current LCA Database Development in Japan – Results of the LCA Project. Int J LCA 9 (6) 355–359
Newman MC, Ownby DR, Mézin LCA, Powell DC, Christensen TRL, Lerberg SB, Anderson B-A, Padma TV (2002): Species Sensitivity Distributions in Ecological Risk Assessment: Distributional Assumptions, Alternate Bootstrap Techniques, and Estimation of Adequate Number of Species. Chapter 7. In: Posthuma L, Suter II GW, Traas TP (eds), Species Sensitivity Distributions in Ecotoxicology. Lewis, Boca Raton, FL, USA, pp 119–132
Norberg J, Swaney DP, Dushoff J, Lin J, Casagrandi R, Levin SA (2001): Phenotypic diversity and ecosystem functioning in changing environments: A theoretical framework. PNAS 98 (20) 11376–11381
Pant R, Van Hoof G, Schowanek D, Feijtel TCJ, de Koning A, Hauschild MZ, Pennington DW, Olsen SI, Rosenbaum R (2004): Comparison between three different LCIA methods for aquatic ecotoxicity and a product Environmental Risk Assessment – Insights from a Detergent Case Study within OMNIITOX. Int J LCA 9 (5) 295–306
Payet J, Larsen HF (2006): Endpoint modelling for the life cycle impact assessment on ecosystems (in preparation)
Payet J (2005): Assessing Toxic Impacts on Aquatic Ecosystems in LCA. Int J LCA 10 (5) 373
Payet J, Jolliet O (2005): Comparative Assessment of the Toxic Impact of Metals on Aquatic Ecosystems: The AMI method. In: Alain Dubreuil (ed), Life-Cycle Assessment of Metals: Issues and research directions. (Proceedings of the International Workshop on Life-Cycle Assessment and Metals. Montreal, Canada. 15–17 April 2002). SETAC, Pensacola, FL, USA
Payet J (2004): Assessing Toxic Impacts on Aquatic Ecosystems in Life Cycle Assessment (LCA). PhD thesis No. 3112 (2004). EPFL, Lausanne, Switzerland
Payet J, Pennington D, Hauschild M, Jolliet O (2002): The AMI method for the assessment of the median impact on ecosystems: Description and guidelines. EPFL, Lausanne, Switzerland
Pennington D, Payet J, Hauschild M (2004): Aquatic ecotoxicological indicators in life-cycle assessment. Environ Toxicol Chem 23 (7) 1796–1807
Posthuma L, Traas TP, Suter II GW (2002a): General Introduction to Species Sensitivity Distribution. Chapter 1. In: Posthuma L, Suter II GW, Traas TP (eds), Species Sensitivity Distribution in Ecotoxicology. Lewis, Boca Raton, FL, USA, pp 3–10
Posthuma L, Suter II GW, Traas TP (2002b): Species Sensitivity Distribution in Ecotoxicology. Lewis, Boca Raton, FL, USA
Pratt JR, Cairns JJr (1996): Ecotoxicology and the Redundancy Problem: Understanding Effects on Community Structure and Function. Chapter 12. In: Newman MC, Jagoe CH (eds), Ecotoxicology – A Hierarchical Treatment. Lewis, Boca Raton, FL, USA, pp 347–370
Rinaman CW, Heil C, Strauss MT, Mascagni M, Sousa M (1996): Probability and Statistics. Chapter 7. In: Zwillinger D (chief), Krantz SG, Rosen KH (eds), Standard Mathematical Tables and Formulae. 30 ed. CRC, Boca Raton, FL, USA
Snell TW, Serra M (2000): Using probability of extinction to evaluate the ecological significance of toxicant effects. Environ Toxicol Chem 19 (9) 2357–2363
Solbe J, Mark U, Buyle B, Guhl W, Hutchinson T, Kloepper-Sams P, Länge R, Munk R, Scholz N, Bontinck, Niessen H (1998): Analysis of the ECETOC Aquatic Toxicity (EAT) database. I – General introduction. Chemosphere 36 (1) 99–113
Straalen NM van, van Leeuwen CJ (2002): European History of Species Sensitivity Distributions. Chapter 3. In: Posthuma L, Suter II GW, Traas TP (eds), Species Sensitivity Distribution in Ecotoxicology. Lewis, Boca Raton, FL, USA, pp. 19–34
Straalen NM van, Denneman CAJ (1989): Ecotoxicological evaluation of soil quality criteria. Ecotox Environ Saf 18, 241–251
Suter II GW (2002): North American History of Species Sensitivity Distributions. Chapter 2. In: Posthuma L, Suter II GW, Traas TP (eds), Species Sensitivity Distribution in Ecotoxicology. Lewis, Boca Raton, FL, USA, pp 11–17
Tanaka Y, Nakanishi J (2000): Mean extinction time of population under toxicant stress and ecological risk assessment. Environ Toxicol Chem 19 (11) 2856–2862
Tilman D (2001): An evolutionary approach to ecosystem functioning. PNAS 98 (20) 10979–10980
Traas TP, van de Meent D, Posthuma L, Hamers T, Kater BJ, de Zwart D, Aldenberg T (2002): The Potentially Affected Fraction as a Measure of Ecological Risk. Chapter 16. In: Posthuma L, Suter II GW, Traas TP (eds), Species Sensitivity Distributions in Ecotoxicology. Lewis, Boca Raton, FL, USA, pp 315–344
Udo de Haes HA, Lindeijer E (2002): The conceptual Structure of Life-Cycle Impact Assessment. Chapter 8. In: Udo de Haes HA, Finnveden G, Goedkoop M, Hauschild M, Hertwich E, Hofstetter P, Jolliet O, Klöpffer W, Krewitt W, Lindeijer E, Müller-Wenk R, Olsen SI, Pennington D, Potting J, Steen B (eds), Life-cycle Impact Assessment: Striving Towards Best Practice. SETAC-Europe, Brussels, Belgium, pp 209–225
Zwart D de (2002): Observed Regularities in Species Sensitivity Distribution for Aquatic Species. Chapter 8. In: Posthuma L, Suter II GW, Traas TP (eds), Species Sensitivity Distributions in Ecotoxicology. Lewis, Boca Raton, FL, USA, pp 315–344

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Appendix A: The PAF approach (OnlineEdition)

The PAF (Potentially Affected Fraction of species) approach is based on the Species Sensitivity Distribution (SSD) which is a statistical distribution describing the variation in the toxicity of a certain substance or mixture among a set of species. This distribution can be of different forms, e.g. triangular, log-logistic, lognormal (all parametric), and it can be made for different types of data (e.g. EC_{50}s or NOECs). It is also possible to avoid the assumption of any distribution function by using non-parametric statistics, i.e. the median combined with e.g. bootstrap technique for confidence limits (Newman et al. 2002). General issues about SSD are most recently described in Posthuma et al. (2002a), Suter II (2002) and in van Straalen and van Leeuwen (2002), which are all introductory chapters in the book 'Species Sensitivity Distribution in Ecotoxicology' (Posthuma et al. 2002b).

The description below covers the single substance PAF, the multi substance PAF (msPAF also called combi-PAF), the 'marginal PAF increase' approach and the most recently proposed approach based on average PAF increase.

A1 Single substance PAF

PAF can be defined as the fraction of species in a (generic) ecosystem/community that is expected to be affected above its no-effect level (i.e. NOEC) or another predefined level of effect (e.g. its EC_{50}) at a given environmental concentration of a toxicant or other stressor (adopted from Traas et al. (2002)).

The PAF approach including the single substance PAF and the multi substance PAF (msPAF, described in Section A2) has most recently been described in details by Traas et al. (2002).

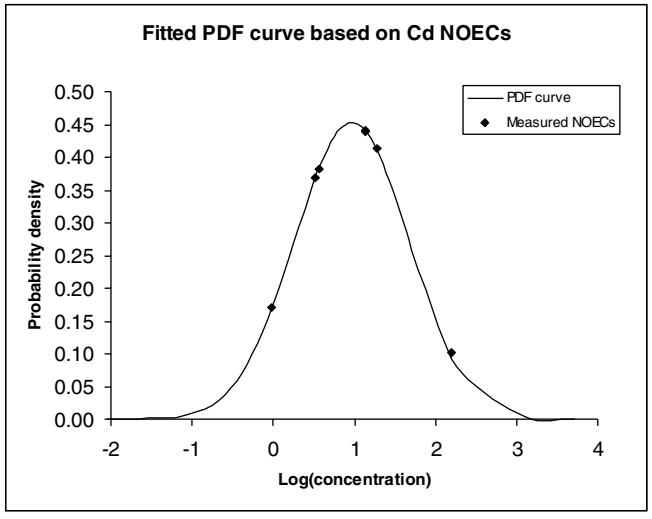

Fig. A1: An example of a fitted Probability Distribution Function (PDF, here a lognormal distribution) based on seven cadmium (Cd) NOECs for different soil species (n = 7, one point hidden in the figure). For the sake of the illustration, the 'NOEC dots' are placed on the curve and not on the x-axis. The log10 sample mean is 0.97 and log10 sample standard deviation is 0.70. The area below the curve to the left of a given NOEC value represents the relative probability (as compared to the total area below the curve) of randomly selecting a species with a lower NOEC value. Data on Cd are taken from Aldenberg et al. (2002) (originally from Straalen & Denneman 1989)

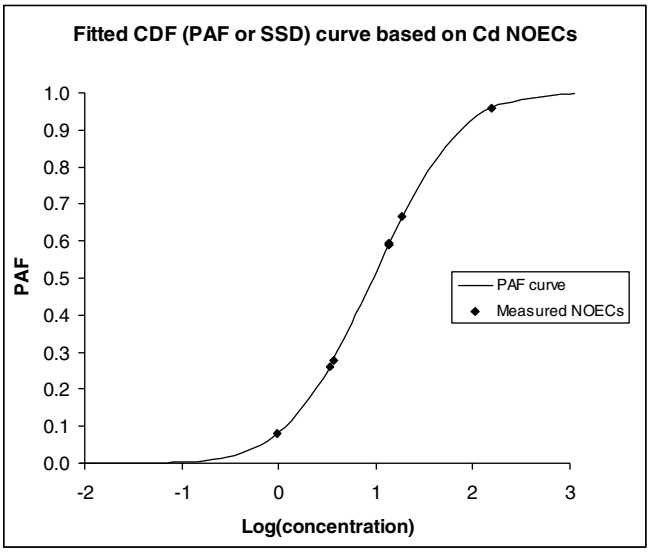

Fig. A2: Fitted lognormal CDF or PAF curve illustrating the relationship between the concentration of a toxicant (here cadmium, Cd) and PAF, i.e. the cumulative probability of exceeding NOECs for a certain fraction of species in the community/ecosystem in question (here a soil community). The dots represent measured values of NOECs for Cd in soil as in Fig. A1. For the sake of the illustration, the 'NOEC dots' are placed on the curve and not on the x-axis. The concentration at which the fraction p of species are having their NOECs (or alternatively, their $L(E)C_{50}$s) exceeded is denoted the hazardous concentration at level p, i.e. HCp. For example the concentration at which 5% (PAF=0.05) of the species are having their NOEC exceeded is denoted HC5. Data on Cd are taken from Aldenberg et al. (2002) (originally from Straalen & Denneman 1989)

The PAF approach is based on the assumption that the distribution of sensitivities of species living in a community (or ecosystem) can be described by a Probability Distribution Function (PDF), e.g. a bell shaped normal distribution, see **Fig. A1**, which after integration yields a Cumulative Distribution Function (CDF) as illustrated in **Fig. A2**.

Whether one uses a SSD function (PDF and CDF) based on a lognormal distribution or a log-logistic distribution is in principle not crucial for the shape of the curves or the outcome of the estimation of the HCp value (or PAF value), as long as the percentile (p value) used is not too low (significantly below 0.05) as shown by van Straalen and van Leeuwen (2002, Fig. 3.3) and Pennington et al. (2004, Fig. 4a and 4b). But of course the input data have to fit the distribution. Otherwise, a non-parametric method having a higher data demand has to be used as described by Newman et al. (2002).

In Fig. A1 and Fig. A2, a lognormal distribution is used as described for the PDF and CDF in, for example, Rinaman et al. (1996, pp 583–584).

The CDF (or PAF curve) equation for a log-logistic distribution is shown below (de Zwart 2002):

$$PAF = \frac{1}{1+e^{-(\log C-\alpha)/\beta}}, \text{ where} \qquad (A1)$$

C : Environmental concentration (of toxicant)

α: Sample mean or location parameter. α is estimated as $\bar{x}$:

$\bar{x} = \frac{1}{n}\sum_{i=1}^{n} x_i$, where n is the number of input log data (x), e.g. log NOECs

β: Scale parameter. β is estimated as β: $\beta = \frac{\sqrt{3}}{\pi}\cdot\sigma$,

where σ is the standard deviation: $\frac{1}{\sqrt{n-1}}\cdot\sum_{i=1}^{n}(x_i-\bar{x})^2$

A2 Multi Substance PAF (msPAF)

One of the advantages of the msPAF approach is that it is able to handle mixtures (hence also called combi-PAF), as described below. The description is mainly based on Traas et al. (2002).

For each of the toxicants in the data set in question, a single-substance PAF is made (in principle as illustrated in the example in Fig. A2). But instead of entering the NOEC values (we assume that it is a NOEC-based SSD) in concentration units (e.g. µg/l), they are expressed in dimensionless hazard units (HUs) by dividing each NOEC value by the parametric median (i.e. the geometric mean) or HC50 of all the NOEC values, done set-by-set for each toxicant. We hereby observe that 1 HU is equal to a concentration where the NOEC is exceeded for 50% of all species tested for all included toxicants. The resulting CDF is pictured with HU on the X-axes and msPAF values on the Y-axes, and the curve type resembles the one in Fig. A2.

If we then want to estimate the potentially affected fraction of species due to a mixture exposure (where we know the environmental concentration of each toxicant), we have to scale each concentration to HUs in the same way as the NOEC values. If all the toxicants in the mixture have the same Toxic Mode of Action (TMoA) (e.g. baseline narcotic), we can use the principles of Concentration Addition (CA) and just add the number of HU. If we assume that the species sensitivity distribution can be described by a log-logistic distribution, then the msPAF can be calculated in the following way:

$$\mathrm{msPAF_{CA}} = \mathrm{PAF_{TMoA}} = \frac{1}{1+e^{-\log(\sum \mathrm{HU_{TMoA}})/\beta_{\mathrm{TMoA}}}} \quad , \qquad (A2)$$

where β_{TMoA} is the average scale parameter (scale factor) over all the toxicants in the mixture.

The scale parameter for each toxicant is calculated as $(\sigma\cdot\sqrt{3})/\pi$, where σ is the standard deviation for each CDF, see Eq. A1.

If the mixture contains toxicants with different TMoAs, the estimations become more complicated. All toxicants with the same TMoAs are treated in groups and, for each group, a PAF_{TMoA} is calculated as described above for concentration addition (see **Eq.** A2). For each single toxicant with different TMoAs (or an unknown TMoA), a PAF is also calculated according to the principles for calculating single substance PAF, see **Eq.** A1. All these PAF values (PAF_{TMoA} and single substance PAF) are then treated by the rules for Response Addition (RA):

$$\mathrm{msPAF_{RA}} = 1 - \prod_{i=1}^{n}(1-\mathrm{PAF_i}) \quad , \qquad (A3)$$

where n is the number of PAF values.

The $msPAF_{RA}$ can in this case be designated $msPAF_{CA+RA}$, because Concentration Addition, CA, precedes Response Addition, RA.

In **Fig. A3**, an example of a msPAF-curve based on HUs is shown.

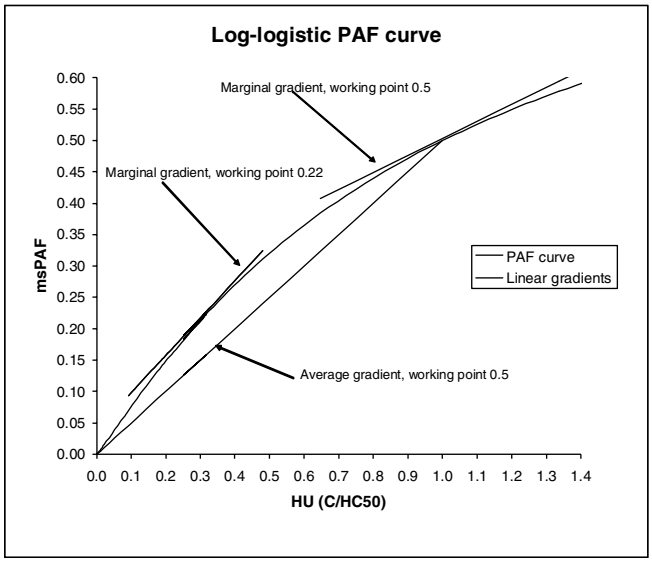

Fig. A3: Log-logistic PAF curve with concentration shown on a linear scale as hazard units, HU (C/HC50). The marginal gradient with working point at PAF = 0.22 and having a slope of 0.59 is the one used in the Eco-indicator 99 method. The marginal gradient and the average gradient, both with a working point at PAF = 0.5, have slopes of 0.27 and 0.50, respectively. The msPAF curve is defined by a β value of 0.4 as used in Eco-indicator 99

A3 'Marginal PAF increase' approach

The 'marginal PAF increase' approach is used in connection with LCIA in the Eco-indicator 99 method (Goedkoop and Spriensma 2001a, 2001b) and most recently described in Huijbregts et al. (2002).

In this approach, the ecotoxicity effect indicator is expressed as the marginal increase in the msPAF, (as compared to the background level msPAF) of the community/ecosystem SSD in question, due to exposure from an emitted toxicant. In other words, the aim is to express the potential impact from

an emitted toxicant by the marginal increase in the number of species having their NOEC values exceeded while taking the background impact (i.e. the number of species already having their NOEC exceeded) into account.

The 'marginal PAF increase' approach is based on the principles of calculating an msPAF as described in Section A2. In the adoption of the approach in Eco-indicator 99 (Goedkoop and Spriensma 2001a, 2001b), it is assumed, because all possible TMoAs are already present in the environment, that an emitted toxicant will imply a concentration addition to a mechanism already present. The method used in Eco-indicator 99 is therefore only based on CA. This assumption may result in an error of above a factor of 100 (Huijbregts et al. 2002). The method is making use of Dutch investigations on SSDs for different compartments (e.g. freshwater) which include measured, existing environmental concentrations of toxicants (mainly pesticides and heavy metals) in support of the calculation of the background msPAF values. Based on these investigations, the Eco-indicator 99 method sets up a general log-logistic msPAF curve (called combi-PAF) with the scaling to HU as described in Section A2. An average β value (scale factor) of 0.4 and a background msPAF value of 22% (geometric mean of msPAF background values for water and soil in the Netherlands) defining the working point is used in this method. The working point is the point on the PAF curve that corresponds to the background impact on the community/ecosystem in question, i.e. the number of HU already present corresponding to a certain msPAF value.

In the 'marginal PAF increase' approach, the increase in PAF due to the emission of a toxicant is not directly calculated from the actual PAF curve (i.e. CDF). To ensure linearity (proportionality) in the calculation and use of the characterisation factor, the slope (slope factor, γ) of the tangent to the curve at the working point is used instead, i.e. a tangential or marginal gradient. The marginal gradient used in the Eco-indicator 99 method (working point at PAF = 0.22) is shown in Fig. A3 together with a marginal gradient and an average gradient (described in Section A4), both at working point 0.5.

In such a marginal approach the PAF increase per HU is dependent on the slope of the tangent (marginal gradient), which is dependent on the location of the working point on the PAF curve as seen from Fig. A3. In Eco-indicator 99, the working point (PAF = 0.22) is defined by the average background impact (background PAF) in the Netherlands as described above. If the working point is placed at PAF = 0.05 instead of PAF = 0.50, the slope is about a factor of 3 higher when β is 0.4, as in the Eco-indicator 99 method. If the β value is set to 0.7 (highest average value within different TMoA (de Zwart 2002)), the slope varies by a factor of 22. Location of the working point can thus have a relatively high impact on the slope depending on the kind of TMoA involved.

In the Eco-indicator 99 method with a working point defined by an msPAF value of 22%, the slope (γ) of the tangent to the generalised msPAF curve in that point is 0.59. The characterisation factor (CF) defined as the marginal increase in msPAF due to a marginal increase in the concentration of a toxicant can be calculated in the following way:

$$CF_{CA} = \mathrm{dmsPAF} = \frac{\gamma}{10^{\alpha}} \cdot \mathrm{dC} \qquad \text{(A4)}$$

Where dC is the concentration increase calculated by the fate model, in this case USES-LCA (Huijbregts et al. 2000), and α is the location parameter (i.e. the $\log_{10}$ sample mean), which can be calculated as the geometric mean (GM) of the NOECs:

$$\mathrm{GM} = \sqrt[1/n]{\prod_{i=1}^{n} x_i} \quad ,$$

where n is the number of NOEC values (x_i) (A5)

So, to be able to calculate the 'effect part' (i.e. $\gamma/10^{\alpha}$) of Eq. A4, only the mean of the NOEC values for the toxicant in question is needed.

The number of NOEC values (n) is specified to 4 in Goedkoop and Spriensma (2001a, 2001b). The minimum requirement of 4 NOEC values (from a minimum of 4 different taxonomic groups) is also used in The Netherlands for an estimation of Environmental Quality Criteria (EQC) by log-logistic probability distribution (de Bruijn et al. 1999, Traas et al. 2002). As stated in Aldenberg et al. (2002) and de Bruijn et al. (1999), it should be tested whether or not the NOEC values fit the probability distribution (i.e. log-logistic) by e.g. Kolmogorov-Smirnov test before this statistical extrapolation method is used.

Huijbregts et al. (2002) have modified the 'marginal PAF increase' approach described above by including the principles of response addition (RA) (see Eq. A3) with the aim of taking different TMoAs into account. They arrive at the following equation for a characterisation factor:

$$CF_{CA+RA} = \mathrm{dmsPAF} = \frac{1-\mathrm{msPAF}}{1-\mathrm{PAF}} \cdot \frac{\gamma}{10^{\alpha}} \cdot \mathrm{dC} \qquad \text{(A6)}$$

where msPAF is the background msPAF (e.g. 0.22 as in Eco-indicator 99 method mentioned above) and PAF is the single substance PAF value for the toxicant in question (calculated according to **Eq.** A1).

A4 'Average PAF increase' approaches

Instead of using a tangential gradient, as in the 'marginal PAF increase' approach, Pennington et al. (2004) argue for using a secant or average gradient, especially at low exposure concentration (below HC5), i.e. in the area where the shape of the CDF curve becomes very uncertain. As shown in Fig. A3, the average gradient is a linear gradient starting

from the chosen working point (e.g. msPAF = 0.5) and having a slope (e.g. 0.5) determined by the working point.

If we assume that the background PAF is below 0.05, but use this value as a working point for an average gradient, the equation becomes (Pennington et al. 2004):

$$CF_{CA} = dmsPAF = \frac{0.05}{(HC5/HC50)/HC50} \cdot dC \qquad (A7)$$

The slope in this case is 0.05/(HC5/HC50).

As recommended by Pennington et al. (2004) and as implemented in the AMI method (Payet 2004, 2005) as a best available practice in LCA, the average gradient may be based on HC50 (working point, PAF = 0.5). In this case, the equation for the characterisation factor becomes:

$$CF_{CA+RA} = dmsPAF = \frac{0.5}{HC50} \cdot dC \qquad (A8)$$

So, in this case, the slope is 0.5 as illustrated in Fig. A3.

As illustrated by the example in **Fig. A4**, the estimation of HC5 (needed in Eq. A7) is much more uncertain than the estimation of HC50.

By choosing the simple average gradient based on HC50 (Eq. A8), a distinction between CA and RA is not necessary – all toxicants can be handled according to CA as shown by Pennington et al. (2004).

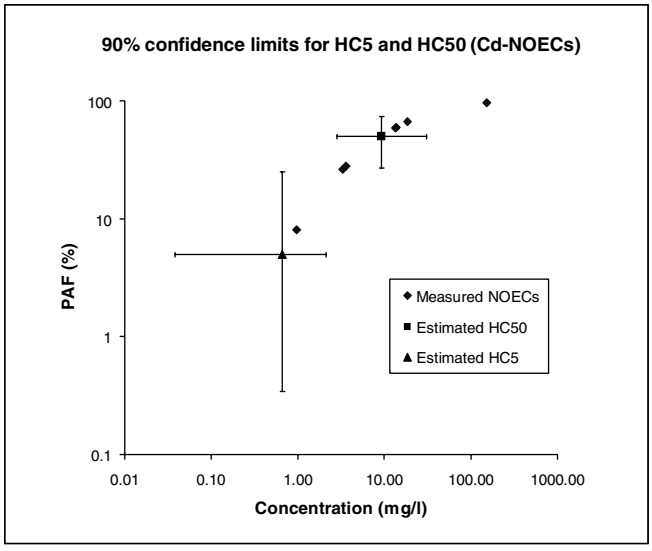

Fig. A4: An example showing the 90% confidence limits (i.e. 5th percentile and the 95th percentile) for both PAF and concentration, for HC50 and HC5 estimated on the basis of seven cadmium (Cd) NOECs for different soil species (n = 7, one point hidden in the figure), fitted to a lognormal distribution. For the sake of the illustration, the NOECs, HC50s and HC5s are not placed on the x-axis, but according to the corresponding PAF-value. The horizontal limits for HC5 are estimated on the basis of the law of extrapolation (Aldenberg et al. 2002, p. 61–62 and Table 5.A1) and the vertical limits for both HC5 and HC50 on the basis of the uncertainty of the fraction affected (Aldenberg et al. 2002, p. 65 and Table 5.A2). The horizontal limits for HC50 are estimated on the basis of student's t-distribution (Campbell 1974, Table A12). Data on Cd are taken from Aldenberg et al. (2002) (originally from Straalen & Denneman 1989)